Studies of the EPR *g* Factors and Hyperfine Structure Constants for Yb^{3+} Ions in Single Crystals of Zircon-structure Orthophosphates

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The EPR *g* factors $g_{\|}$ and $g_{\perp}$ of Yb^{3+} and the hyperfine structure constants $A_{\|}$ and $A_{\perp}$ of $^{171}Yb^{3+}$ and $^{173}Yb^{3+}$ in crystals of the zircon-structure orthophosphates YPO_4, $LuPO_4$, and $ScPO_4$ are calculated from the calculation formulas of the EPR parameters for a $4f^{13}$ ion in tetragonal symmetry. In these formulas, the contributions to the EPR parameters from the *J*-mixing between the ground $^2F_{7/2}$ and excited $^2F_{5/2}$ states, the admixtures between the lowest Kramers doublet $\Gamma\gamma$ and the other 6 Kramers doublets (or irreducible representations) via the crystal-field and orbital angular momentum (or hyperfine structure) interactions and the covalency reduction effect are included. The calculated values agree with the observed ones. In the calculations, the superposition model is used to estimate the crystal-field parameters. It is found intrinsic parameters $\bar{A}(R_0)$ that the superposition model with the same R_0 for Yb^{3+} in various zircon-type compounds obtained by considering local lattice relaxation are not scattered as those obtained by using the structural data of pure zircon-type compounds. It appears that, in order to obtain suitable superposition model parameters, the local lattice relaxation should be taken in to account.

Key words: Electron Paramagnetic Resonance; Crystal-field Theory; Superposition Model; Yb^{3+}; YPO_4; $LuPO_4$; $ScPO_4$.

1. Introduction

Introducing rare-earth ions into crystals can subtly alter the properties of materials and leads to applications in optical and electronic devices. Among them the oxides with zircon-structure (i. e., $ZrSiO_4$ type) are useful host materials to study the properties and applications [1 – 3]. Many studies of optical and EPR spectra for rare-earth ions in zircon-type compounds have appeared [1 – 6]. For example, the EPR *g* factors $g_{\|}$, $g_{\perp}$, of Yb^{3+} and hyperfine structure constants $A_{\|}$, $A_{\perp}$ for the isotopes $^{171}Yb^{3+}$ and $^{173}Yb^{3+}$ in zircon-structure orthophosphates YPO_4, $LuPO_4$ and $ScPO_4$ crystals were measured [6]. However, no theoretical studies related to the structural data of Yb^{3+} centers have been made for these EPR parameters. Since zircon-type compounds have tetragonal structure [3], in this paper, we first of all establish the calculation formulas for the lowest Kramers doublet Γ_6 or Γ_7 of the ground $^2F_{7/2}$ state of a $4f^{13}$ ion in tetragonal symmetry. In these formulas, the contributions to the EPR parameters from the *J*-mixing between the ground $^2F_{7/2}$ and excited $^2F_{5/2}$ states via crystal field interaction, the admixture among the other $(7-1=6)$ Kramers doublets (or irreducible representations) Γ_x and the ground Γ_6 or Γ_7 doublet via the crystal-field and orbital angular momentum (or hyperfine structure) interactions and the covalency reduction effect are considered. From these formulas, the EPR parameters for Yb^{3+} in various zircon-structure orthophosphates are calculated. In the calculations, the tetragonal crystal-field parameters are estimated from the superposition model [7] and the structural data of Yb^{3+} centers. The results (including the local lattice relaxation and superposition model parameters) are discussed.

2. Calculations

Yb^{3+} has a $4f^{13}$ electronic configuration, a $^2F_{7/2}$free ion ground state and a $^2F_{5/2}$ excited state. The tetragonal crystal-field splits the $^2\mathrm{F}_{7/2}$ and $^2\mathrm{F}_{5/2}$ states into four and three Kramers doublets, respectively. The lowest lying (or ground) doublet is Γ_6 or Γ_7, corresponding to the average $\bar{g} \approx 2.667$ or 3.429 to the first order [8]. Because of the crystal-field J-mixing between $J = 7/2$ and $J = 5/2$ states, the basis function of the ground doublet $\Gamma\gamma$ (Γ_6 or Γ_7) can be obtained by diagonalizing a 14×14 energy matrix for the $4\mathrm{f}^{13}$ ion in tetragonal symmetry. Thus, we have

$$\begin{aligned}|\Gamma\gamma(\mathrm{or}\,\gamma')\rangle &= \sum_{M_{J1}} C(^2\mathrm{F}_{7/2};\Gamma\gamma(\mathrm{or}\,\gamma')M_{J1})|^2\mathrm{F}_{7/2}M_{J1}\rangle \\ &\quad + \sum_{M_{J2}} C(^2\mathrm{F}_{5/2};\Gamma\gamma(\mathrm{or}\,\gamma')M_{J2})|^2\mathrm{F}_{5/2}M_{J2}\rangle,\end{aligned} \tag{1}$$

where the subscripts γ and γ' denote the two components of the Γ irreducible representation. M_{J1} and M_{J2} are half-integers in the ranges $-7/2$ to $7/2$ and $-5/2$ to $5/2$, respectively.

Considering that the other $(4+3-1=6)$ Kramers doublets (or irreducible representations) Γx (which are obtained by diagonalizing the 14×14 energy matrix) may mix with the ground $\Gamma\gamma$ doublet via the crystal-field interaction H_{CF} and orbital angular momentum $\hat{L}$ (or hyperfine structure equivalent operator $\hat{N}$), and so make the contributions to the EPR parameters, the calculation formulas of the EPR parameters for the $4\mathrm{f}^{13}$ ion in tetragonal symmetry should include the second-order contribution. Thus, these formulas can be expressed as

$$g_\parallel = g_\parallel^{(1)} + g_\parallel^{(2)}, \quad g_\perp = g_\perp^{(1)} + g_\perp^{(2)}, \tag{2}$$

$$\begin{aligned} g_\parallel^{(1)} &= 2g_J(^{2S+1}L_J)\langle\Gamma\gamma|\mathbf{J}_z|\Gamma\gamma\rangle \\ &= 2\Big\{\sum_{M_{J1}} g_J(^2\mathrm{F}_{7/2})|C(^2\mathrm{F}_{7/2};\Gamma\gamma M_{J1})|^2 M_{J1} \\ &\quad + \sum_{M_{J2}} g_J(^2\mathrm{F}_{5/2})|C(^2\mathrm{F}_{5/2};\Gamma\gamma M_{J2})|^2 M_{J2}\Big\} \\ &\quad + 4\sum_{M_{J2}} g'_J(^2\mathrm{F}_{7/2},{}^2\mathrm{F}_{5/2})\left[(5/2+1)^2 - M_{J2}^2\right]^{1/2} \\ &\quad \cdot C(^2\mathrm{F}_{7/2};\Gamma\gamma M_{J2})C(^2\mathrm{F}_{5/2};\Gamma\gamma M_{J2}),\end{aligned}$$

$$g_\parallel^{(2)} = 2{\sum_X}' \frac{\langle\Gamma\gamma|\hat{H}_{\mathrm{CF}}|\Gamma_X\gamma_X\rangle\langle\Gamma_X\gamma_X|\hat{J}_Z|\Gamma\gamma\rangle}{E(\Gamma_X) - E(\Gamma)},$$

$$\begin{aligned} g_\perp^{(1)} &= 2g_J(^{2\mathrm{S}+1}\mathrm{L}_J)\langle\Gamma\gamma|\mathbf{J}_x|\Gamma\gamma'\rangle \\ &= \sum_{M_{J1}}(-1)^{7/2-M_{J1}+1}\left[7\cdot 9/4 - (M_{J1}-1)M_{J1}\right]^{1/2} \\ &\quad \cdot g_J(^2\mathrm{F}_{7/2})C(^2\mathrm{F}_{7/2};\Gamma\gamma M_{J1}) \\ &\quad \cdot C(^2\mathrm{F}_{7/2};\Gamma\gamma' M_{J1}-1), \\ &\quad + \sum_{M_{J2}}(-1)^{5/2-M_{J2}+1}\left[5\cdot 7/4 - (M_{J2}-1)M_{J2}\right]^{1/2} \\ &\quad \cdot g_J(^2\mathrm{F}_{5/2})C(^2\mathrm{F}_{5/2};\Gamma\gamma M_{J2}) \\ &\quad \cdot C(^2\mathrm{F}_{5/2};\Gamma\gamma' M_{J2}-1), \\ &\quad + 2\sum_{M_{J2}} g'_J(^2\mathrm{F}_{5/2},{}^2\mathrm{F}_{5/2}) \\ &\quad \cdot \left[(5/2+M_{J2}+1)(5/2+M_{J2}+2)\right]^{1/2} \\ &\quad \cdot C(^2\mathrm{F}_{7/2};\Gamma\gamma M_{J2})C(^2\mathrm{F}_{5/2};\Gamma\gamma' M_{J2}-1),\end{aligned}$$

$$g_\perp^{(2)} = 0,$$

$$A_\parallel = A_\parallel^{(1)} + A_\parallel^{(2)}, \quad A_\perp = A_\perp^{(1)} + A_\perp^{(2)}, \tag{3}$$

$$\begin{aligned} A_\parallel^{(1)} &= 2PN_J(^{2S+1}L_J)\langle\Gamma\gamma|\mathbf{N}_z|\Gamma\gamma\rangle \\ &= P\Big\{\sum_{M_J1} N_J(^2\mathrm{F}_{7/2})|C(^2\mathrm{F}_{7/2};\Gamma\gamma M_{J1})|^2 M_{J1} \\ &\quad + \sum_{M_J2} N_J(^2\mathrm{F}_{5/2})|C(^2\mathrm{F}_{5/2};\Gamma\gamma M_{J2})|^2 M_{J2}\Big\} \\ &\quad + 2P\sum_{M_{J2}} N'_J(^2\mathrm{F}_{7/2},{}^2\mathrm{F}_{5/2})\left[(5/2+1)^2 - M_{J2}^2\right]^{1/2} \\ &\quad \cdot C(^2\mathrm{F}_{7/2};\Gamma\gamma M_{J2})C(^2\mathrm{F}_{5/2};\Gamma\gamma M_{J2}),\end{aligned}$$

$$A_\parallel^{(2)} \approx 2P\sum X' \frac{\langle\Gamma\gamma|\hat{H}_{\mathrm{CF}}|\Gamma_X\gamma_X\rangle\langle\Gamma_X\gamma_X|\hat{N}_{\mathrm{Z}}|\Gamma\gamma\rangle}{E(\Gamma_X) - E(\Gamma)}$$

$$\begin{aligned} A_\perp^{(1)} &= 2PN_J(^{2S+1}L_J)\langle\Gamma\gamma|\mathbf{J}_{\mathrm{x}}|\Gamma\gamma'\rangle \\ &= \sum_{M_{J1}}(-1)^{15/2-M_{J1}+1}\left[15\cdot 17/4 - (M_{J1}-1)M_{J1}\right]^{1/2} \\ &\quad \cdot N_J(^4\mathrm{I}_{15/2})C(^4\mathrm{I}_{15/2};\Gamma\gamma M_{J1})\end{aligned}$$

$$
\begin{aligned}
&\cdot C({}^4\mathrm{I}_{15/2};\Gamma\gamma' M_{J1}-1)\\
&+\sum_{M_{J2}}(-1)^{132-M_{J2}+1}\left[13\cdot 15/4-(M_{J2}-1)M_{J2}\right]^{1/2}\\
&\qquad\cdot N_J({}^4\mathrm{I}_{13/2})C({}^4\mathrm{I}_{13/2};\Gamma\gamma M_{J2})\\
&\qquad\cdot C({}^4\mathrm{I}_{13/2};\Gamma\gamma' M_{J2}-1)\\
&+2\sum_{M_{J2}}g'_J({}^2\mathrm{F}_{5/2},{}^2\mathrm{F}_{5/2})\\
&\qquad\cdot\left[(\frac{5}{2}+M_{J2}+1)(\frac{5}{2}+M_{J2}+2)\right]^{1/2}\\
&\qquad\cdot C({}^2\mathrm{F}_{7/2};\Gamma\gamma M_{J2})C({}^2\mathrm{F}_{5/2};\Gamma\gamma' M_{J2}-1),
\end{aligned}
$$

$$A_\perp^{(2)}=0,$$

where the parameters g_J, g'_J, N_J and N'_J [note: g'_J and N'_J occur in the expansions of (2) and (3)] for various states can be obtained from [8] and [9]. P is the dipolar hyperfine structure constant. In the above formulas, the second-order terms $g_\perp^{(2)}$ and $A_\perp^{(2)}$ vanish because none of the six Γx nonzero matrix elements with ground $\Gamma\gamma$ for both H_{CF} and x or y component of $\hat{L}$ and $\hat{N}$ operators.

For the studied zircon-type crystals R(R=Y, Lu, Sc)PO_4:Yb^{3+}, the average values of $\bar{g}$ [6] suggest that the ground doublet is Γ_6. In these crystals, Yb^{3+} ions replace the R^{3+} ions, and they occupy the sites having noncentrosymmetrical $\mathrm{D}_{2\mathrm{d}}$ point symmetry. So, the Yb^{3+} ion is surrounded by a tetragonal dodecahedral array of eight neighboring oxygens-four of these are at one distance R_1 and the other four at a slightly different distance R_2. The structural parameters R_i and θ_i (θ_i is the angle between the R_i and C_4 axis) [10] of the studied zircon-type compounds RYO_4 are shown in Table 1. The crystal field Hamiltonian in the tetragonal symmetry can be expressed as

$$
\begin{aligned}
\hat{H}_{\mathrm{CF}}=&B_2^0C_0^2+B_4^0C_0^4+B_6^0C_0^6\\
&+B_4^4(C_4^4+C_{-4}^4)+B_6^4(C_4^6+C_{-4}^6),
\end{aligned}
\tag{4}
$$

where the B_k^q are the crystal-field parameters which also occur in the above 16×16 energy matrix.

Table 1. Structure data for Yb^{3+} in zircon-structure orthophosphates.

Compounds	R_1 (Å)		R_2 (Å)		θ_1	θ_2
	I[a]	II[b]	I[a]	II[b]		
YPO_4	2.313	2.296	2.374	2.357	103.67°	30.22°
LuPO_4	2.264	2.268	2.346	2.350	103.47°	30.95°
ScPO_4	2.150	2.213	2.277	2.340	103.17°	31.62°

[a] Values in pure crystals given in [10]. [b] Values estimated by considering the local lattice relaxation.

Table 2. the orbital reduction factor k and intrinsic parameters $\bar{A}_k(R_0)$ with $R_0=2.343$ Å for Yb^{3+} in zircon-structure orthophosphates.

		$\bar{A}_2(R_0)$ (cm^{-1})		$\bar{A}_4(R_0)$ (cm^{-1})		$\bar{A}_6(R_0)$ (cm^{-1})	
	k	I[a]	II[b]	I[a]	II[b]	I[a]	II[b]
YPO_4	0.941	385	285	40	33.2	22.5	22.5
LuPO_4	0.949	385	285	37.3	34.3	27	27
ScPO_4	0.941	185	285	29.3	28.2	27	27

[a] Estimated from the structural data of pure crystals.
[b] Estimated by considering the local lattice relaxation.

From the superposition model [7], the crystal-field parameters can be written as

$$B_k^q=\sum_{j=1}^{n}\bar{A}_k(R_0)(R_0/R_j)^{t_k}K_k^q(\theta_j,\phi_j),\tag{5}$$

where the coordination factor $K_k^q(\theta_j,\phi_j)$ can be obtained from the local structural data of the studied system. $\bar{A}_k(R_0)$ are the intrinsic parameters with the reference distance R_0, t_k are the power law exponents. For the $(\mathrm{YbO}_8)^{13-}$ group, the above superposition model parameters were not reported. We estimate them as follows: the exponents $t_2=7$, $t_4=12$ and $t_6=11$ are taken as those obtained in the similar trivalent rare-earth ion Er^{3+} in zircon-type compounds [3] and the parameters $\bar{A}_k(R_0)$ with $R_0=2.343$ are the adjustable parameters obtained by fitting the calculated EPR parameters ($g_\parallel$, $g_\perp$, $A_\parallel$ and $A_\perp$) to the observed values. In the calculations, the free-ion values of the spin-orbit coupling coefficient $\zeta_{4\mathrm{f}}^0(\approx 2950\ \mathrm{cm}^{-1}$ [8]) and the dipolar hyperfine structure constant P_0 ($\approx 388.4(7)\times 10^{-4}\ \mathrm{cm}^{-1}$ and $106.5(2)\times 10^{-4}\ \mathrm{cm}^{-1}$ for the isotopes ${}^{171}\mathrm{Yb}^{3+}$ and ${}^{173}\mathrm{Yb}^{3+}$, respectively [8]) should be multiplied by the orbital reduction factor k (which is also taken as an adjustable parameter) because of the covalency effect for the Yb^{3+} ion in crystals. Thus, by fitting the calculated EPR parameters $g_\parallel$, $g_\perp$, $A_\parallel$ and $A_\perp$ based on the structural data of host RPO_4 crystals to the observed values, the parameters $\bar{A}_k(R_0)$ and the factor k

Table 3. EPR *g* factors and hyperfine structure constants (in 10^{-4} cm^{-1}) for Yb^{3+} in YPO_4, $LuPO_4$ and $ScPO_4$ crystals (data in parentheses are errors).

	$g_{\parallel}$			$g_{\perp}$			$A_{\parallel}$			$A_{\perp}$			Isotope
	Cal.[a]	Cal.[b]	Expt.[6]	Cal.[a]	Cal.[b]	Expt.[6]	Cal.[a]	Cal.[b]	Expt.[6]	Cal.[a]	Cal.[b]	Expt.[6]	
YPO_4	1.492	1.584	1.526(1)	3.152	3.122	3.120(3)	381.8(6)	405.8(7)	408(4)	820.5(14)	813.5(13)	824(7)	$^{171}Yb^{3+}$
							104.5(1)	111.2(1)	111(1)	224.6(4)	222.7(4)	235(2)	$^{173}Yb^{3+}$
$LuPO_4$	1.361	1.387	1.338(10)	3.265	3.253	3.233(3)	347.2(6)	354.5(6)	360(3)	851.1(14)	847.9(14)	853(7)	$^{171}Yb^{3+}$
							95.2(2)	97.3(2)	97(1)	233.4(4)	232.5(4)	243(3)	$^{173}Yb^{3+}$
$ScPO_4$	0.965	0.973	0.973(2)	3.410	3.411	3.405(3)	240.9(4)	245.6(4)	263(3)	889.4(15)	889.2(15)	897(7)	$^{171}Yb^{3+}$
							66.1(1)	67.4(1)	70(1)	243.9(4)	243.8(4)	257(1)	$^{173}Yb^{3+}$

[a] Calculated from the structural data of host crystals. [b] Calculated by considering the local lattice relaxation.

for RPO_4:Yb^{3+} can be obtained. They are shown in Table 2. The comparisons between the calculated and experimental EPR parameters for RPO_4:Yb^{3+} are shown in Table 3.

From Table 2 one can find that the intrinsic parameters $\bar{A}_4(R_0)$ and $\bar{A}_6(R_0)$ for Yb^{3+} in various zircon-structure orthophosphates are close to one another, however the $\bar{A}_2(R_0)$ for Yb^{3+} in $ScPO_4$ is much smaller than those in YPO_4 and $LuPO_4$. It is astonishing that the values of $\bar{A}_2(R_0)$ for Yb^{3+} in $ScPO_4$ are so different from those in similar orthophosphates. This may, in our opinion, be due to the neglect of local lattice relaxation caused by introducing the impurity ion Yb^{3+}. We note that for RPO_4:Yb^{3+} the ionic radius $r_i(\approx 0.858$ Å [11]) of the impurity Yb^{3+} is close to those of the replaced Y^{3+} $r_h(\approx 0.893$ Å [11]) and $Lu^{3+}(\approx 0.85$ Å [11]), but it is larger than the radius $r_h(\approx 0.732$ Å [11]). Considering the local lattice relaxation due to the size mismatch, we can reasonably assume that the angles θ_i are unchanged and estimate the impurity-ligand distances R_i by the empirical formula [12, 13]

$$R = R_H + (r_i - r_h)/2, \qquad (6)$$

where R_H is the corresponding distance in the pure crystal. Thus we can estimate the Yb^{3+}–O^{2-} distances R_1 and R_2 for Yb^{3+} in various zircon-structure orthophosphates. They are also shown in Table 1.

By using these local structural data in above formulas and fitting the calculated EPR parameters to the observed values, the intrinsic parameters $\bar{A}_K(R_0)$ for RPO_4:Yb^{3+} are obtained and collected in Table 2. The comparisons between the calculated and experimental EPR parameters are also shown in Table 3.

3. Discussion

From Table 2 it can be found that the intrinsic parameters $\bar{A}_K(R_0)$ of the superposition model [in particular $\bar{A}_2(R_0)$] for Yb^{3+} in various zircon-structure orthophosphates obtained by studying EPR data from the local structural data are not as scattered as those obtained from the structural data of the pure crystals. So, these parameters $\bar{A}_K(R_0)$ are more suitable in physics, and our above opinion about local lattice relaxation is reasonable. It appears that in order to obtain reasonable superposition model parameters from EPR data, the lattice relaxation caused by the substitution of the impurity for the host ion in crystals should be taken in account.

The calculated EPR parameters $g_{\parallel}$, $g_{\perp}$ for Yb^{3+} and $A_{\parallel}$, $A_{\perp}$ for the isotopes $^{171}Yb^{3+}$ and $^{173}Yb^{3+}$ in various zircon-structure orthophosphates agree with the observed values, suggesting that the theoretical method is reasonable. In the calculations, we find that for the parameters $g_{\parallel}$ and $A_{\parallel}$, the contributions due to the second-order perturbation terms is the same in sign and about 15% in magnitude, compared with the corresponding contribution from the first-order term. For instance, $g_{\parallel}^{(1)} \approx 1.374$ and $g_{\parallel}^{(2)} \approx 0.210$ for YPO_4:Yb^{3+}; $A_{\parallel}^{(1)} \approx 351.6 \times 10^{-4} cm^{-1}$ and $A_{\parallel}^{(2)} \approx 52.2 \times 10^{-4} cm^{-1}$ for the isotope $^{171}Yb^{3+}$, and $A_{\parallel}^{(1)} \approx 96.2 \times 10^{-4} cm^{-1}$ and $A_{\parallel}^{(2)} \approx 14.8 \times 10^{-4} cm^{-1}$ for the isotope $^{173}Yb^{3+}$ in YPO_4. So, to explain more reasonably the EPR parameters of Yb^{3+} in crystals, the second-order perturbation contributions should be included.

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